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IN THE UNITED STATES PATENT AND TRADEMARK OFFICE  
BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

APPEAL BRIEF FOR THE APPELLANTS

Ex parte KIM et al.

METHOD FOR MANUFACTURING CATALYTIC OXIDE ANODE USING HIGH  
TEMPERATURE SINTERING

Serial Number: 10/022,357  
Filed: December 20, 2001  
Appeal No.:  
Group Art Unit: 1763  
Examiner: R. Culbert

Submitted herewith is an Appeal Brief. A check in the amount of Three Hundred and Forty Dollars (\$340.00) is enclosed to cover the official fees for the Appeal Brief. Please charge any fee deficiencies required with respect to this paper, or overpayment to our Deposit Account No. 01-2300, **referencing docket number 101190-00022.**

Respectfully submitted,

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Date: December 6, 2004



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

BEFORE THE BOARD OF PATENT APPEALS AND INTERFERENCES

In re the Application of:

Kwang-Hook KIM et al.

Examiner: R. Culbert

Application No.: 10/022,357

Group Art Unit: 1763

Filed: December 20, 2001

Attorney Dkt. No.: 101190-00022

For: METHOD FOR MANUFACTURING CATALYTIC OXIDE ANODE USING HIGH  
TEMPERATURE SINTERING

**BRIEF ON APPEAL**

Date: December 6, 2004

This is an appeal from the action of the Examiner dated April 6, 2004, finally rejecting claim 2, the only claim currently pending in this application, as being unpatentable over certain prior art under 35 U.S.C. § 103. A Notice of Appeal was timely filed on October 6, 2004 with a Petition for Extension of Time. This Brief is being timely filed.

**I. REAL PARTY IN INTEREST**

The real parties in interest in the present application on appeal are Korea Atomic Energy Research Institute and Technology Winners Co., LTD.

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## II. RELATED APPEALS AND INTERFERENCES

There are no related appeals or interferences known to the appellant, appellant's representative or assignee that will directly affect or be directly affected by or have a bearing on the Board's decision in the pending appeal.

## III. STATUS OF CLAIMS

Claim 2, the only claim pending in the present application is being appealed.

## IV. STATUS OF AMENDMENTS

All amendments have been entered.

## V. SUMMARY OF THE CLAIMED SUBJECT MATTER

The claimed subject matter of independent claim 2 relates to a method for manufacturing a catalytic oxide anode using high temperature sintering, wherein a TiO<sub>2</sub>-screening layer, which is a metal oxide layer of TiO<sub>2</sub>, SnO<sub>2</sub>, RuO<sub>2</sub>, or IrO<sub>2</sub>, sintered at 450 to 550°C, is added between titanium support and a surface of the oxide anode, coated with a precursor solution of RuCl<sub>3</sub> or IrCl<sub>3</sub> in hydrochloric acid according to a brushing or dipping method, dried at 60°C for 10 min, thermally treated at 250 to 350°C for 10 min, and finally sintered at 600 to 700°C for 1 to 2 hours, said TiO<sub>2</sub>-screening layer serving as an valve metal oxide for preventing the activity of the anode from being lowered owing to the oxidation of a titanium base metal caused upon sintering of the anode at high temperature and the solid diffusion of an oxide into the anode surface,

said valve metal oxide being selected from the group consisting of  $\text{TiO}_2$ ,  $\text{SnO}_2$ ,  $\text{RuO}_2$ , and  $\text{IrO}_2$  sintered at 450 to 550°C.

The present specification explains that “[a]ccording to the present invention, a decomposition efficiency of organic substances by the oxide anode is increased by 50 to 100% because the oxide anode is manufactured at 600 to 700°C, which is higher than a conventional sintering temperature range for manufacturing  $\text{RuO}_2$  or  $\text{IrO}_2$  anode ... thereby the performance of the catalytic oxide anode is improved” (page 11, lines 7-14, of the present specification).

## VI. GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL

Claim 2 was rejected under 35 U.S.C. § 103(a) as being unpatentable over Beer (British Patent No. 1,480,807).

## VII. ARGUMENT

### **Rejection of claim 2 under 35 U.S.C. 103(a) (Beer)**

#### i. The Law

1. The law regarding factual inquiries to determine obviousness/non-obviousness.

Several basic factual inquiries must be made to determine obviousness or non-obviousness of patent application claims under 35 U.S.C. § 103. These factual inquiries are set forth in Graham v. John Deere Co., 383 U.S. 1,17,148 U.S.P.Q. 459, 467 (1996):

Under § 103, the scope and content of the prior art are to be determined; differences between the prior art and the claims at issue are to be ascertained; the level of ordinary skill in the pertinent art resolved. Against this backdrop, the obviousness or non-obviousness of the subject matter is determined.

The specific factual inquiries set forth in *Graham* have not been considered or properly applied by the Examiner formulating the rejections of claims 4 and 5. Particularly the differences between the prior art and the claims were not properly determined. As stated by the Federal Circuit in In re Ochiai, 37 U.S.P.Q. 2d 1127, 1131 (Fed. Cir. 1995):

[t]he test of obviousness *vel non* is statutory. It requires that one compare the claim's subject matter as a whole with a prior art to which the subject matter pertains. 35 U.S.C. § 103.

The inquiry is highly fact-specific by design.... When the references cited by the Examiner fail to establish a *prima facie* case of obviousness, the rejection is improper and will be overturned. In re Fine, 837 F.2d 1071, 1074, 5 U.S.P.Q. 2d 1596, 1598 (Fed. Cir. 1988). (Emphasis added.)

When rejecting claims under 35 U.S.C. § 103, an Examiner bears an initial burden of presenting a *prima facie* case of obviousness. A *prima facie* case of obviousness is established only if the teachings of the prior art would have suggested the claimed subject matter to a person of ordinary skill in the art. If an Examiner fails to establish a *prima facie* case, the rejection is improper and will be overturned. See: In re Rijckaert, 9 F.3d 1531, 28 U.S.P.Q. 2d. 1955 (Fed. Cir. 1993). “If examination.... does not produce a *prima facie* case of unpatentability, then without more the applicant is entitled to the grant of the patent.” In re Oetiker, 977 F.2d 1443, 1445-1446 24 U.S.P.Q. 2d. 1443, 1444 (Fed. Cir. 1992).

Appellants respectfully submit that the Examiner has not made a proper *prima facie* rejection under 35 U.S.C. § 103(a), because the Beer reference cited fails to teach or suggest the invention of present claim 2.

In the Office Action dated April 6, 2004, claim 2 was rejected under 35 U.S.C. § 103(a) as being unpatentable over Beer (British Patent Specification No. 1,480,807).

The April 6, 2004, Office Action indicated that a sintering temperature range and a layer material are the same as those of Beer, and a TiO<sub>2</sub>-screening layer is formed

between a ruthenium oxide formed while being in contact with a titanium surface and a platinum metal oxide, and thus, a  $\text{TiO}_2$ -screening layer according to Beer may be used with the same aim as the subject invention.

However, Appellants respectfully submit that claim 2 of the subject invention would not have been obvious in view of Beer.

Appellants first note that the subject matter of pending claim 2 requires sintering at two different temperatures, namely both sintering at  $450^\circ\text{C}$  to  $550^\circ\text{C}$  **and** at  $600^\circ\text{C}$  to  $700^\circ\text{C}$ . As Beer, at best, only very broadly mentions a single oxide formation and adherence step at  $400^\circ\text{C}$ - $650^\circ\text{C}$ , present claim 2 would not have been obvious over Beer for this reason alone.

Appellants further note that, electrode production prior art, including Beer, suggest a sintering temperature of  $400$ - $550^\circ\text{C}$  or  $400$ - $650^\circ\text{C}$  (Beer) to oxidize a coating solution, but in the course of practically producing the electrode, the sintering temperature does not exceed  $550^\circ\text{C}$ . The reason for this is that, in the case of conducting high temperature sintering at  $600^\circ\text{C}$  or higher, since a base metal (or valve metal) is simultaneously oxidized in conjunction with oxidation of a precursor solution, coated on the base metal to form a desired oxide on the base metal, a base metal oxide ( $\text{TiO}_2$ ). That is to say,  $\text{TiO}_2$  is solid-diffused to a surface of the desired oxide, thereby occupying a surface of the electrode, resulting in reduced electrode activity, and performances of the electrode are significantly reduced because surface resistance of the electrode is increased, and the oxide coated on the base metal is easily stripped from the base metal.

Unlike the indication in the Office Action, prior art, including Beer, disclose that the sintering of the oxide is conducted at 400-650°C, but do not mention that the performances of the electrode are unexpectedly improved when the sintering is conducted at 600°C or higher (refer to FIGS. 3 and 4).

However, conventional patents or documents describe characteristics of the high temperature sintering, but do not overcome problems, such as increased surface resistance of the electrode, reduced activity of the electrode, and the stripping of the oxide, caused by the diffusion of the base metal oxide ( $\text{TiO}_2$ ) onto an electrode surface because of the oxidation of the base metal when the solution coated on the surface of the base metal is sintered at high temperatures.

In other words, the precursor solution ( $\text{RuCl}_3$ ,  $\text{IrCl}_3$  and the like) coated on the surface of the base metal is more sufficiently oxidized during the high temperature sintering than at a conventional sintering temperature, thereby providing improved characteristics to the oxide of the electrode surface.

However, this advantage is completely countervailed by the oxidation of the base metal during the high temperature sintering and the solid diffusion of the base metal into the surface of the oxide (hence, in practice, sintering is usually conducted at temperatures less than 550°C even though Beer's final sintering temperature is within a range of 400-650°C). Moreover, in order to prevent the characteristics of the electrode from being reduced, before the precursor solution ( $\text{RuCl}_3$ ,  $\text{IrCl}_3$  and the like) coated on the surface of the base metal is sintered at high temperatures, a metal oxide layer, such as  $\text{TiO}_2$  (it assures a predetermined electric conductivity when it is sintered at 550°C),  $\text{SnO}_2$ ,  $\text{IrO}_2$ , or  $\text{RuO}_2$ , is formed on the surface of the base metal to form another electric



conductive oxide layer at 450-550°C, for example, a metal-like oxide layer, on the surface of the base metal, and the solution, which is capable of producing the desired oxide according to a procedure similar to a conventional oxide production process, is coated on the metal oxide layer, dried at 60-70°C, and heat treated at 250-350°C.

This procedure is repeated to increase the number of coated layers, and the resulting structure is finally sintered at 600-700°C to produce the desired oxide layer. In this regard, Beer does not mention any electric conductive oxide layer formed at 450-550°C, that is, the  $\text{TiO}_2$  (base metal oxide)-screening layer for preventing the oxidation of the base metal and the solid diffusion of the base metal onto the surface of the desired oxide layer during the final sintering (i.e., metal oxide for preventing the oxidation of base metal and the diffusion of the base metal onto the desired oxide of  $\text{IrO}_2$  or  $\text{RuO}_2$  during high temperature sintering).

The additionally applied oxide layer serves to reduce the oxidation of the base metal (Ti) during the high temperature sintering for formation of the final surface oxide, thereby maintaining a high concentration of the desired oxide in the final oxide layer unlike the case of employing no additional oxide layer. Accordingly, it can be seen that oxidation characteristics of organics are improved when the  $\text{TiO}_2$  (base metal oxide)-screening layer is employed during high temperature sintering.

Although the Examiner asserted that "a  $\text{TiO}_2$ -screening layer is formed between a ruthenium oxide formed while being in contact with a titanium surface and a platinum metal oxide, and thus, a  $\text{TiO}_2$ -screening layer according to Beer may be used with the same aim as the subject invention", Beer does not mention the  $\text{TiO}_2$ -screening layer. The three-stage sintering process described in Beer includes a drying stage at 80-

120°C, a moisture removal and crystallization stage at 175-300°C, and an oxide formation and attachment stage at 400-600°C, which is intended to maintain a multicoated layer to enlarge a layered structure during when the precursor solution ( $\text{RuCl}_3$  or  $\text{IrCl}_3$  in solution) is coated in a multilayer coating manner, and to reduce a thermal shock caused by a physicochemical phenomenon during the sintering, and this process is adopted to form the desired oxide layer after the  $\text{TiO}_2$ -screening layer is formed.

When a coating solution ( $\text{RuCl}_3$  or  $\text{IrCl}_3$  in solution) is coated in the multilayer coating manner, after the first coating is conducted using the coating solution, water and diluent (the coating solution is an acid or alcohol base solution) are evaporated from the coating solution at 80-120°C and 175-300°C, and metal ions are bonded to oxygen in air to start to form oxide crystals while a Cl component is dissociated.

If the coating solution is additionally applied on the previous layer while this stage is omitted, since the previous layer is not completely dried and set, when the coating solution is additionally painted on the previous layer, the additional coating solution is mixed with the previous insufficiently dried layer, and thus, a laminate having distinct layers cannot be formed.

Furthermore, if the coating solution is heated to 400-600°C using a furnace immediately after it is applied on the coated layer, since the base metal and oxide have different thermal expansion coefficients, the coating solution insufficiently adheres to the previous layer and is easily separated from the base metal, and many cracks are formed on the final oxide layer, and thus, the surface becomes unstable. As well, the case of sintering the coating solution directly applied on the Ti base metal is different

from the case of forming the TiO<sub>2</sub>-screening layer acting as the firm oxide layer between the Ti base metal and the desired oxide layer according to the subject invention in view of the diffusion of the oxidized base metal onto the desired oxide layer when the base metal is sintered at high temperatures.

In other words, in the case of employing no TiO<sub>2</sub>-screening layer, when the solution coated on the surface of the base metal is oxidized by heat, the base metal is heated and oxidized. The base metal oxide is easily diffused from the surface of the base metal to the oxide layer of the electrode surface, and thus, many base metal oxides are present on the electrode surface. However, in the case of employing the TiO<sub>2</sub>-screening layer, since the firm oxide layer is formed between the base metal and the desired coated layer to be sintered, the base metal oxide is not easily moved to the surface of the desired coated oxide layer. Accordingly, in the case of employing the TiO<sub>2</sub>-screening layer, when a metal oxide concentration is analyzed according to a depth of the oxide layer, the base metal component content is low in the final oxide layer.

According to the invention of present claim 2, the oxide layer, such as TiO<sub>2</sub>-screening layer, positioned under the final oxide layer, functions to suppress the oxidation of the base metal and the solid diffusion of the base metal to the surface of the oxide layer during the high temperature sintering, and to reduce a concentration gradient of the oxide between the base metal layer and the desired oxide layer, thereby preventing the final oxide layer from being stripped.

Thus, the subject matter of pending claim 2, which requires sintering at two different temperatures, namely both sintering at 450°C to 550°C **and** at 600°C to 700°C,

would not have been obvious over Beer, which, at best, only very broadly mentions a single oxide formation and adherence step at 400°C-650°C.

Thus, as claimed elements of present claim 2, including sintering at two different temperatures and the advantages achieved thereby, are missing from Beer, Appellants respectfully submit that the presently claimed invention would not have been obvious over Beer.

Additionally, as described in the present specification, "[v]ariables affecting electrochemical and physical properties of the oxide anode include an etching method of titanium base metal, a coating method of metal chloride which is coated on the base metal, the number of a coating process, and a sintering temperature. Among the above variables, the sintering temperature is the most important, and it has been defined within a range from 400 to 550°C during the manufacture of RuO<sub>2</sub> or Ir O<sub>2</sub> anodes. The sintering temperature is defined within a range from 400 to 550°C so that the oxide anode has a sufficient anode activity, and low resistance of its surface when RuCl<sub>3</sub> or IrCl<sub>3</sub> used as a coating material of the oxide anode is converted to RuO<sub>2</sub> or IrO<sub>2</sub>" (see page 3, lines 12-24, of the present specification, underlining added).

The present specification further explains that "[a]ccording to the present invention, a decomposition efficiency of organic substances by the oxide anode is increased by 50 to 100% because the oxide anode is manufactured at 600 to 700°C, which is higher than a conventional sintering temperature range for manufacturing RuO<sub>2</sub> or IrO<sub>2</sub> anode ... thereby the performance of the catalytic oxide anode is improved" (page 11, lines 7-14, of the present specification).

Thus, the present specification demonstrates unexpected results and advantages when the catalytic oxide anode is sintered within the range 600°C or to 700°C.

Although Beer mentions a generic phase of heat treatment of "heating of a temperature ranging from 400° to 650° C "(page 2, lines 12-13), Beer nowhere teaches or suggests the unexpected advantages achieved by sintering within the specific range 600°C to 700°C, as compared to sintering at a lower temperature alone. Furthermore, Beer provides no examples of heat treating within the within the specific range 600°C to 700°C. All of the examples in Beer are directed to a relatively short heat treatment (15 minutes to 50 minutes) at a lower temperature that would not achieve the unexpected results and advantages achieved by the presently claimed invention including a step of sintering at a temperature in the specific range of 600°C to 700°C for 1 to 2 hours.

Thus, Appellants submit that the unexpected results and advantages achieved by the presently claimed invention, including a step of sintering at a temperature in the specific range of 600°C to 700°C for 1 to 2 hours thereby, would not have been obvious over Beer, which nowhere teaches or suggests that such advantages can be achieved at a temperature within the species of 600°C to 700°C. Appellants submit that the temperature species of 600°C to 700°C, with the unexpected results achieved therein, is patentably distinct from the partially overlapping temperature range of 400° to 650° C of Beer.

Thus, as the unexpected results achieved by the invention of present claim 2 are missing from Beer, Appellants respectfully submit that the presently claimed invention would not have been obvious over Beer for this additional reason.

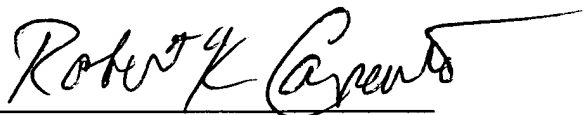
### Conclusion

For all of the above-noted reasons, it is strongly contended that clear differences exist between the present invention as recited in claim 2 and the prior art relied upon by the Office Action.

This final rejection being in error, therefore it is respectfully requested that this Honorable Board of Patent Appeals and Interferences reverse the Examiner's decision in this case and indicate the allowability of claim 2.

In the event that this paper is not considered timely filed, Appellants respectfully petition for an appropriate extension of time. Any fees for such extension, together with any additional fees which may be due with respect to this paper, may be charged to Deposit Account No. 01-2300, making reference to attorney docket number 101190-00022.

Respectfully submitted,

A handwritten signature in black ink, reading "Robert K. Carpenter", with a long horizontal flourish extending to the right.

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## APPENDIX 1

### CLAIMS ON APPEAL

1. (Canceled)

2. (Previously Presented) A method for manufacturing a catalytic oxide anode using high temperature sintering, wherein a  $\text{TiO}_2$ -screening layer, which is a metal oxide layer of  $\text{TiO}_2$ ,  $\text{SnO}_2$ ,  $\text{RuO}_2$ , or  $\text{IrO}_2$ , sintered at 450 to 550°C, is added between titanium support and a surface of the oxide anode, coated with a precursor solution of  $\text{RuCl}_3$  or  $\text{IrCl}_3$  in hydrochloric acid according to a brushing or dipping method, dried at 60°C for 10 min, thermally treated at 250 to 350°C for 10 min, and finally sintered at 600 to 700°C for 1 to 2 hours, said  $\text{TiO}_2$ -screening layer serving as an valve metal oxide for preventing the activity of the anode from being lowered owing to the oxidation of a titanium base metal caused upon sintering of the anode at high temperature and the solid diffusion of an oxide into the anode surface, said valve metal oxide being selected from the group consisting of  $\text{TiO}_2$ ,  $\text{SnO}_2$ ,  $\text{RuO}_2$ , and  $\text{IrO}_2$  sintered at 450 to 550°C.